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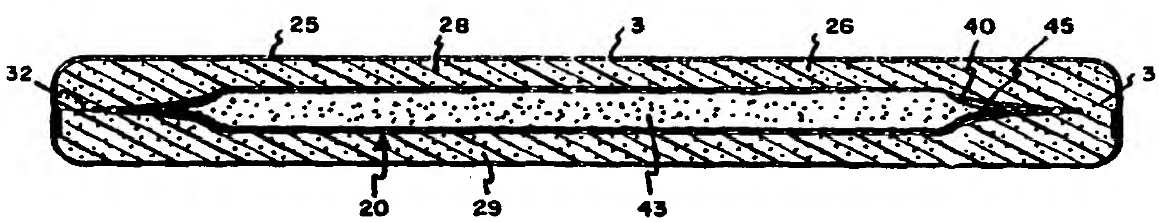

(57) Abstract

A powder thermal storage composition activated by exposure to microwave energy including a combination of a powder, a phase-change material, and a microwave-sensitive material combined to provide an impregnated powder (43) that has at least more than 50 n % to substantially all of the individual powder particles impregnated with both the phase-change material and the microwave-sensitive material; the impregnated powder (43) may be produced by the process of: (I) mixing an emulsion with a powder; (II) mixing a powder first with either microwave-sensitive material or phase-change material and then mixing the resultant powder with the phase-change material or the microwave-sensitive material, respectively; or (III) vigorously mixing the powder while spraying microwave-sensitive material and phase-change material (either separately or as a mixture) into the vigorously mixing powder; the thermal storage mixture (43) may be used in a container, to provide a thermal construction comprising a seat cushion (3) having a thermal storage unit (20) therein.

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MICROWAVE-ACTIVATED POWDER THERMAL STORAGE COMPOSITION; AND METHOD

The present application is related to United States patent application Serial No. 08/438,443, to Gideon Salee, having the same filing date as the present application, the text of which is incorporated by reference as if completely rewritten herein.

Field of the Invention

The present invention relates to microwave-activated thermal storage compositions for thermal storage units. In particular, the invention concerns arrangements which are activated or heated through exposure to microwave energy and which possess significant thermal storage ability and provide an advantageously slow release of heat over an extended period of time. Principles according to the present invention may be used, for example, in thermal seat cushions, foot and hand warmers, body warmers, and as warmth extenders for foods. Also according to the present invention, methods relating to preparation and use of such arrangements are presented. Typically, the invention includes materials where free-flowing powders are obtained. The powders are impregnated such that more than at least 50 percent (population or number, hereinafter referred to as n%), preferably at least 75 n%, and most preferably substantially all of the individual powder particles have both phase-change materials and microwave-sensitive materials adsorbed and/or absorbed therewith.

Background of the Invention

Thermal storage arrangements (whether heated by exposure to microwave energy or otherwise) have been widely used for a variety of purposes. Those which are activated by microwave energy have been used for various heating applications. One such arrangement, US 5,211,949 to Salyer, uses as a thermal storage material a silica powder with a phase-change material. The material is claimed to be a free-flowing powder when

the particle size is between 0.007 and 0.07 microns in diameter above and below the melting temperature of the phase-change material.

Other arrangements where materials have been combined with a powder include US 4,008,170 to Allen, where silica powder has been combined with up to 9 parts by weight water. The water/silica powder is used for cooling or as a source of water.

In US 4,253,983, Blanie discloses paraffin compositions having improved heat reservoir properties. The disclosed paraffin compositions include paraffin combined with two different fillers so as to improve thermal conductivity and plasticity.

The present invention provides for a simple system for providing stored heat that has improved heat transfer characteristics when heated with microwaves.

Summary of the Invention

The invention typically includes a thermal storage composition activatable by exposure to microwave energy, the composition including a powder wherein more than at least 50 n% of all particles of the powder are impregnated with both a phase-change material and a microwave-sensitive material. Preferably at least 75 n%, and most preferably substantially all, of the particles of the powder are impregnated with both the phase-change materials and the microwave-sensitive materials. The thermal storage composition is a free-flowing impregnated powder. As discussed below, the phase-change material is typically a microwave-insensitive material. The microwave-sensitive material preferably is a low volatility, microwave-sensitive glycol while the phase-change material preferably is an organic wax material. For uses as a seat cushion the solid phase-change material preferably includes material having a melting point of at least 30°C and most preferably a melting point within the range of 35°C-65°C. A preferred thermal storage composition includes a powder of calcium silicate, a phase-change material of paraffin wax and a microwave-sensitive material selected from the group consisting of a microwave-sensitive glycol, water or a mixture thereof. Most preferably, the powder is calcium silicate, the phase-change material is paraffin wax, and the microwave-sensitive material is selected from the group consisting of

dipropylene glycol, diethylene glycol, higher oligomers of propylene or ethylene glycol, water, or mixtures thereof.

Another embodiment of the invention includes a thermal storage composition activatable by exposure to microwave energy, the composition including a powder, an emulsion of a microwave-sensitive material, and a phase-change material, wherein the powder is impregnated with the emulsion. For uses that include microwave heating it is preferred that more than at least 50 n% of all particles of the powder are impregnated with both a phase-change material and a microwave-sensitive material. More preferably, at least 75 n%, and most preferably, substantially all, of the particles of the powder are impregnated with both the phase-change materials and the microwave-sensitive materials. Typically the product powders will be an impregnated powder that is free flowing at temperatures of use.

In another embodiment of the invention, the thermal storage compositions are typically a product of a process that includes emulsifying a phase-change material and a microwave-sensitive material; and impregnating a powder with the emulsion in an amount effective to form a free-flowing impregnated powder.

In yet another embodiment of the invention, a process typically used for making thermal storage compositions includes the steps of emulsifying a mixture of a phase-change material and a microwave-sensitive material; and mixing the emulsion with a powder able to be impregnated with the emulsion in an amount wherein the thermal storage composition is a free-flowing impregnated powder.

In a yet further embodiment of the invention there is provided a process for producing a thermal storage composition including the steps of mixing a microwave-sensitive material or a phase-change material with a powder able to be impregnated with the materials, in an amount, at a temperature, and at a mixing shear rate effective to impregnate the microwave-sensitive material or the phase-change material with the powder particles to form a partially impregnated powder able to receive additional microwave-sensitive material or phase-change material; and mixing the microwave-sensitive or phase-change material not selected in the preceding step with the partially impregnated powder in an amount, at a temperature, and at a mixing shear rate

effective to impregnate the microwave-sensitive or the phase-change material with the partially impregnated particles, wherein a product is obtained with more than at least 50 n% of the particles having both the microwave-sensitive material and the phase-change material impregnated therewith. Preferably at least 75 n%, and most preferably substantially all, of the powder particles have both the microwave-sensitive material and the phase-change material impregnated therewith.

A further embodiment of the invention provides for a process for producing a thermal storage composition by the steps of vigorously mixing a powder; spraying microwave-sensitive material and phase-change material, either separately or as a mixture, into the vigorously mixing powder in a manner effective to form an impregnated free-flowing powder wherein more than at least more than 50 n%, preferably at least 75% to substantially all, powder particles have both said microwave-sensitive material and said phase-change material impregnated therewith.

Another embodiment of the invention provides for a thermal storage unit which is activated by exposure to microwave energy, where the thermal storage unit includes a microwave-transparent container; and, a thermal storage composition enclosed within the container; the thermal storage composition including a powder impregnated with a microwave-sensitive material and a phase-change material, wherein at least more than 50 n% of all particles of the powder are impregnated with both a phase-change material and a microwave-sensitive material. Preferably the microwave-sensitive material is water, the phase-change material is paraffin wax, and the container is a flexible pouch. Preferably, the impregnated powder was impregnated with an emulsion and at least 75 n% to substantially all particles of the powder are impregnated with both a phase-change material and a microwave-sensitive material.

A further embodiment of the invention provides for a heating construction that includes a microwave-transparent outer cover; and, a thermal storage unit activatable by exposure to microwave energy; the thermal storage unit being positioned within the outer cover; the thermal storage unit including a microwave-transparent outer pouch and an impregnated powder composition within the outer pouch; the impregnated powder composition including a powder impregnated with a microwave-sensitive material and

a phase-change material, wherein at least more than 50 n% of all particles of the powder are impregnated with both a phase-change material and a microwave-sensitive material. Preferably the microwave-sensitive material is water; and the phase-change material is paraffin wax and the impregnated powder was impregnated with an emulsion. The microwave-transparent outer cover typically is a seat cushion.

A yet further embodiment of the invention includes a process of storing thermal energy for release over a extended period of time; the process including the step of exposing a powder composition to microwave energy; the powder composition being a powder; and, a microwave-sensitive material and a phase-change material impregnated with the powder, wherein at least more than 50 n% of all particles of the powder are impregnated with both the phase-change material and the microwave-sensitive material; the step of exposing the powder composition to microwave energy including exposing the powder composition to microwave energy of appropriate power and for a sufficient period of time to heat the powder composition to a temperature above the melting point of the phase-change material and to melt the phase-change material. Preferably at least 75 n% to substantially all of the particles of the powder are impregnated with both the phase-change materials and the microwave-sensitive materials.

Further details of the above summarized embodiments are provided below.

Brief Description of the Drawings

Fig. 1 is a perspective view of a cushion according to the present invention.

Fig. 2 is a top plan view of a thermal storage unit according to the present invention which may, for example, be used in the arrangement of Fig. 1.

Fig. 3 is a cross-sectional view along line 3-3 of Fig. 1.

Detailed Description of the Invention

The principles of the present invention, with respect to thermal storage units and arrangements, may be applied in a variety of systems. The systems may be used, for example, as sources of warmth for humans, for example, as seat cushions, shoe inserts, foot warmers, hand warmers, etc. The principles of the present invention may, however,

be applied in a variety of other systems as well, for example, to provide a thermal blanket for food containers or other items to be kept warm. In the figures, a thermal storage unit according to the present invention is shown in the embodiment of a seat cushion. From the general principles described herein, application in a variety of other arrangements will be apparent.

Referenced numeral 1, Fig. 1, generally designates an arrangement or heating construction according to the present invention, including a thermal storage unit therein. Arrangement 1, Fig. 1, generally comprises a flexible seat cushion 3. The cushion 3 depicted is sufficiently flexible to be folded, and includes carrying handles 5 and 6, comprising, for example, polypropylene webbing or similar material. A closure arrangement to retain the cushion folded, when desired, is indicated generally at 9. The closure arrangement 9 depicted comprises a strap 10 including a first member 11 of a hook and loop closure system thereon. A second member 12 of the hook and loop closure system is indicated on, and underneath, the underside of the cushion 3, at 12. Thus, when cushion 3 is folded, strap 10 can be folded to engage section 11 with 12 to retain the cushion 3 in a closed orientation. The hook and loop closure system may comprise, for example, the well-known system sold under the mark VELCRO®. Alternative closure arrangements, for example, snaps, buckles, buttons or ties, may, of course, be used.

Cushion 3 includes therein, according to the present invention, a thermal storage unit 20, Fig. 3. In operation, cushion 3 is exposed to microwave energy. After several minutes of exposure to the microwave energy, for example, in a microwave oven, the cushion 3 will become relatively warm (a temperature of the thermal storage unit of around 35°C-60°C being typical). This effect is accomplished because cushion 3 includes, embedded therein, a thermal storage unit 20 according to the present invention.

In Fig. 3, a cross-sectional view of cushion 3 is depicted. Cushion 3 includes an outer cover or sheath 25, which encloses the various inner components. The sheath 25 may comprise a fabric, for example, a fire retardant nylon or, more preferably, a polyolefin. The inner components include foam pad 26 which surrounds thermal storage unit 20. More specifically, in the embodiment shown, foam pad 26 includes first and

second pads or walls 28 and 29, with thermal storage unit 20 positioned therebetween. Along edge 31, walls 28 and 29 may be integral with one another, i.e., they may be merged continuously with no interface or seam. Along at least part of edge 32, on the other hand, walls 28 and 29 can be separated, for insertion of thermal storage unit 20 during assembly. In preferred embodiments, foam pad 26 comprises a sheet of foam slit to form an envelope, such that thermal storage unit 20 can be readily inserted therein during assembly. Alternatively, foam pad 26 comprises a sheet of foam folded, or foam pad 26 may comprise two pads to form a sandwich type construction such that thermal storage unit 20 can be readily inserted therein during assembly.

Still referring to Fig. 3, thermal storage unit 20 comprises an outer pouch 40 having sealed therein a thermal storage composition comprising an impregnated powder 43. The powder useful for impregnation most preferably comprises a powder such as a calcium silicate powder. In the preferred embodiment the powder is impregnated with an emulsion of a microwave-sensitive fluid and phase-change material so that more than at least 50 n%, and most preferably 75 n% to substantially all, of the individual powder particles are impregnated with both materials. Also, the impregnated powder should be free flowing at conditions of use.

As used herein the term "impregnation" includes the adsorption and/or absorption of microwave-sensitive and phase-change materials to powder particles. Similarly, the term "adsorb" includes the known phenomena of "adsorption" and "absorption" of liquid with the powder particles. This is because either one or both phenomena may be present in any particular impregnated powder particle; that is, the same particle may have material adsorbed to its surface as well as have material absorbed within pores or spaces of the particle. Further, not wishing to be bound by the particular explanation of what is occurring in terms of absorption or absorption of the materials to the powders, the important consideration is that the phase-change materials and the microwave-sensitive materials are sufficiently "held" by the powder that a free-flowing impregnated powder is obtained at the temperatures of intended use.

A top plan view of the thermal storage unit 20 is depicted in Fig. 2. For the embodiment shown in Fig. 2, thermal storage unit 20 includes outer pouch 40, and a

second inner pouch 45. That is, outer pouch 40 completely encloses inner pouch 45. Alternately stated. thermal storage unit 20 can be a "double bag" arrangement, with outer pouch 40 sealing a second inner pouch 45 for security. This is advantageous because material received within inner pouch 45 will, upon exposure to microwave energy, become relatively hot, and the material enclosed within inner pouch 45 is a free-flowing powder. Thus, should seals in inner pouch 45 fail, or should a failure or puncture occur in inner pouch 45, outer pouch 40 will inhibit leakage of powder. An important consideration for the inner and outer pouches 40 and 45 is that the pouch material melts at temperatures in excess of those produced on heating the powder in a microwave oven. A high melt temperature is more important than in the embodiment revealed in the copending parent application, Serial No. 08/124,931, now United States Patent No. 5,424,519, because higher localized temperatures are expected to be encountered with the powder used herein than with the liquid version.

For the arrangement shown in Fig. 2, outer pouch 40 comprises a film folded along fold line 47 with inner pouch 45 positioned therein. Edge seals 48 may comprise a variety of types of seals, for example, heat seals or adhesive seals. For the preferred arrangement described and shown, preferably outer pouch 40 is formed from a heat sealable film, and edge seals 48 comprise heat seals, formed in a conventional manner. Optional inner pouch 45 also preferably comprises a heat-sealable film. For the arrangement shown in Fig. 2, inner pouch 45, not shown in detail, includes peripheral heat seals 49.

In a further embodiment, the inner pouch 45 can be dispensed with and the impregnated powder 43 freely distributed in the inner space of the outer pouch 40. In this embodiment, the impregnated powder 43 would be held in place by the first or outer pouch 40. This arrangement is also acceptable as the impregnated powder 43 will not escape as readily as a liquid would. In this respect, in a yet further embodiment of the invention, the inner pouch 45 can be of a gas permeable material. Gas permeability would allow volatile components to escape should the thermal storage unit be overheated in the microwave oven by exposing it to microwave radiation at high power for too long a period. As such, this further embodiment offers a significant safety improvement as

overheating would tend to produce hot gases that could cause an impermeable pouch to burst should the internal pressures become too high. This possibility is present especially with the embodiment where the components in the pouch are relatively low boiling liquids such as water. On the other hand, initially water and similar volatile materials will contribute to more even distribution of temperature in the system by mass transfer, i.e., volatilization at hot spots and condensation in cooler areas.

In an embodiment where the microwave-sensitive material is water, it is expected that the volatile gas driven off on overheating to be water vapor. A permeable sheath and pouch cover would allow water vapor driven off the powder by overheating to escape. Then, ambient levels of water vapor in the environment can penetrate to the impregnated powder 43 and allow the powder to readsorb any water driven off by excess heating. This latter point is especially true for calcium silicate which will readily adsorb ambient water vapor from the air. Thus, in this respect, a preferred powder material has at least some hygroscopic characteristics for absorbing water from the atmosphere.

Referring now to Fig. 3, as previously-indicated, impregnated powder 43 is sealed within either (1) both outer pouch 40 and inner pouch 45, or (2) only outer pouch 40. In preferred embodiments, impregnated powder 43 provides three basic operations in use: (1) it is a free-flowing powder and formable, and thus, is comfortable to the user of the cushion 3; (2) the impregnated powder 43 easily spreads over a greater surface area if desired; and (3) it has good thermal characteristics such as low levels of hot spots and acceptable heat transfer from the microwave-sensitive material to the phase-change material, on heating in a microwave oven.

The impregnated powder 43 comprises a mixture of materials to serve two primary purposes in a preferred manner: it is readily heated up upon exposure to microwave energy; and, it will dissipate heat therefrom, in cooling back to ambient, at least in part by a phase transfer of a material impregnated in the powder. By "readily heated up upon exposure to microwave energy" in this context, is meant that it will heat to a selected or preferred temperature, for example, typically to at least 30°C to 30°C-90°C, and usually to a selected point within the range of about 30°C-65°C, upon

exposure to microwave energy in a 700 to 1000 watt microwave oven, for a period of 3 to 15 minutes or so.

Preferred operation of the thermal storage unit 20 turns upon use of preferred materials. Details with respect to these materials are as follows.

1. The Emulsion

Emulsions, used in preparing thermal storage compositions according to one embodiment of the present invention, comprise a form of liquid/solid-in-solid mixture, for example a "wax-in-liquid" or an "oil-in-liquid" emulsion wherein the wax or oil phase, at 25°C, is solid; and in which the wax or oil phase, after the arrangement has been sufficiently heated up upon exposure to microwave energy, becomes a liquid. Herein the terms "wax", "oil" and "phase-change material" are used interchangeably to refer to a liquid phase which is at least initially dispersed with another liquid phase (microwave-sensitive material). In typical applications, the "oil phase" or "phase-change material" in the mixture should comprise a material having a melting point somewhere within the range of about O°C to 100°C, more preferably 30°C-90°C, and most preferably 35°C-65°C. The emulsion formed can be either a phase-change material in microwave-sensitive material emulsion or a microwave-sensitive material in phase-change material emulsion. The type of emulsion is determined by the particular application. Preferably, the phase-change material does not absorb microwave energy. Although some microwave absorption may occur by the phase-change material, it is preferred that the microwave-sensitive material be a significantly more efficient absorber of microwave energy than the phase-change material. However, under some conditions, as discussed further below, it may be advantageous to have a small amount of material present that is both a phase-change and a microwave-sensitive material.

Principal characteristics to be considered in selecting the material for the phase-change material concern the following. The melting point of the phase-change material should be a melting point at a temperature satisfactory for the use to which the thermal storage unit is to be placed. For example, if the thermal storage composition is used as a part of thermal storage unit for a seat cushion (for example, in an arrangement

as shown in Figs. 1-3), it will be preferred that the phase-change material be chosen to have a melting point of at least 40°C, preferably about 50°C-60°C, typically and most preferably at around 50°C-55°C. Such a temperature is relatively warm, but suitable for an internal storage unit within a seat cushion.

On the other hand, if the thermal storage composition is being used in a system more likely to come into closer contact with exposed skin, and relatively sensitive areas of the body, it may be desirable to use as the "oil phase" a material having a lower melting point. For example, if the thermal storage composition is used as part of thermal storage unit for a hand warmer, or foot warmer, for bare hands or bare feet, a melting point on the order of about 30°C-40°C may be preferred, because higher temperatures may be uncomfortable for the user. On the other hand the thermal storage composition may be used for its coolant effects as it warms to ambient temperature and cools the surroundings (e.g., medical cooling packs).

Other factors to be considered in the selection of the material for the phase-change material of the emulsion or mixture, concern heat of fusion (or latent heat of melting). In general, the greater the heat of fusion, the greater the amount of heat given off, per unit weight, as the phase-change material cools and solidifies; and thus, the better the phase-change material will operate in a thermal storage composition, because, after having been heated to liquid, a longer period of time, with greater energy loss, will be achieved before the phase-change material returns to a solid. Alternatively, the greater the heat of fusion, the more energy or heat the phase-change material will be able to store per unit weight during the initial heating up in the microwave oven.

Another factor to be considered in selecting the phase-change material is the ability to form a stable mixture (for example, an emulsion) with the liquid phase (microwave-sensitive material). In general, materials that can relatively readily form an emulsion which is stable over a temperature range on the order of about 0°C-90°C, will be preferred. It is anticipated that, for reasons stated hereinbelow, at temperatures of typical use (e.g., room temperature or cool outdoor temperatures), the typical microwave-sensitive material will be water; thus, the ability to form an emulsion with water is generally an issue. Preferred is the microwave-sensitive material (e.g., water)

in phase-change material emulsion (e.g., wax) so that small amounts of water can be used with consequent large amounts of wax.

Paraffin wax is a preferred material for use in compositions for thermal storage units according to the present invention, especially when the thermal storage unit is used as part of a seat cushion. Paraffin wax is a mixture of solid hydrocarbons having the general formula C_nC_{2n+2} and typically has a melting point somewhere within the range of 45°C-60°C. Particular waxes can be selected for preferred melting points. Thus, a particular paraffin wax can be selected and obtained with a melting point of about 50°C-57°C, i.e., 53°C, if desired. Paraffin waxes in general exhibit melting points over the preferred ranges for use with systems according to the present invention; they readily form emulsions with liquid phases such as water; they are relatively nontoxic; they exhibit desirable heat-storage capabilities; and they are relatively inexpensive and easy to obtain. It is foreseen that derivatives of paraffin wax, such as alkylated paraffins, having similar melting point ranges, may also be used. The term "paraffin wax" as used herein, is meant to include not only materials according to the general formula C_nH_{2n+2}, but also modified waxes such as alkylated paraffin wax derivatives of such materials and similar compounds. Examples include long chain alcohols (e.g., stearyl alcohol and cetyl alcohol), long chain saturated fatty acids (e.g., capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, and myristic acid) and the like.

Other usable materials in the "oil phase" and systems according to the present invention include: beeswax; (mp. 62°C-65°C); candelilla wax (mp. 68°C-70°C); carnauba wax (mp. 82°C-85.5°C); cotton wax; wool wax; montan wax (mp. 80°C-86°C); and, mixtures of waxes. A preferred paraffin wax for systems according to the present invention is a fully refined paraffin having a nominal melting point of 53°C.

The preferred microwave-sensitive phase of the emulsion should comprise a material which readily absorbs microwave energy and converts the microwave energy to heat and, as a result, rapidly heats. Preferably, water or water mixtures (such as water/alcohol, water/glycol, or water/glycerol) are used. Water, of course, is also desirable from the point of view of toxicity and cost. In some systems, aqueous solutions of various salts may be desirable for the microwave-sensitive phase, for example,

solutions of calcium chloride, sodium chloride, aluminum sulfate, magnesium bromide or magnesium sulfate. The presence of such salts in the aqueous phase will modify the freezing point of the liquid microwave-sensitive material, its heat capacity, and the ability to form the emulsion.

The emulsifier or surfactant may comprise any surface active agent effective, and in an amount effective, to achieve sufficient dispersion of one phase in the other, typically by reducing the surface tension of the oil or wax and providing the phase-change material in sufficiently small droplets for the emulsion to form. The particular surfactant used may depend upon the phase-change material and liquid microwave-sensitive material chosen. Many conventional surfactants for mixing oil and aqueous phases can typically be used. When the solid phase comprises paraffin wax and the liquid microwave-sensitive material comprises water, acetylene-base surfactants, such as the Surfinol®s available from duPont (Wilmington, DE, USA), are useful. Other useable surfactants include alkanol amides; alkylaryl sulfonates; amine acetates; amine oxides; sulfonated amines and amides; block polymers; carboxylated alcohol ethoxylates; ethoxylated alkyl phenols; ethoxylated amines and amides; ethoxylated fatty acids; ethoxylated fatty esters and oils; fatty esters; glycerol esters; glycol esters; imidazoline(s); isothionates; lanolin-based surfactants; lecithin(s); olefin sulfonates; phosphate esters; organic phosphorous derivatives; polymeric surfactants (polysaccharides, acrylic acids or acrylamides); propoxylated or ethoxylated fatty acids; propoxylated or ethoxylated alcohols or alkylphenols; soaps; sorbitol derivatives; sulfates and sulfonates of ethoxy alkyl phenols; sulfates and sulfonates of oils and fatty acids; sulfates of alcohols and ethoxylated alcohols; sulfonated benzene, toluene or xylene; and, sulfonates of dodecylbenzene, tridecylbenzene, alkylnaphthalene(s), and petroleum.

In general, the amount of surfactant used should be an amount effective to achieve formation of a stable emulsion for the length of time necessary to ship, store and adsorb the material on the powder. From the specific example of the seat cushion described hereinbelow, a typical formulation will be understood.

The emulsion used to impregnate the powder may comprise a mixture, by weight percent (hereinafter wt%), as follows: paraffin wax 45-95 wt% (melting point about

55°C); surfactant (Surfinol®) 0-12 wt%; water 5-45 wt%; preservative, Ucracide 250 (a glutaraldehyde) available from Union Carbide, 0.1-0.2 wt%; and Proxel GLX (1,2-benzisothiazolin-3-one), 0.1-0.2 wt%. Preferably, the paraffin wax should have a small amount of BHT mixed therewith in order to inhibit oxidation; an amount not in excess of 1 wt% (0.1 to 1 wt%) of the paraffin wax is preferred.

The paraffin wax (molten) can be, and preferably will have been, blended with microwave-sensitive material such as water (and if desired surfactant, etc.), by high shear mixing, to provide a particle size distribution (by number) in the resulting emulsion effective to impregnate and obtain the powder discussed herein. If desired, the emulsion can be filtered, for removal of large particulate material, before it is used to impregnate the powder.

The total amount of emulsion used will be an amount sufficient to coat the surface and/or fill the pores of the powder particles within the internal pouch yet provide a free-flowing impregnated powder at the temperature of operation. The emulsions may include adjuvants such as: surfactants or emulsifiers, preservatives and/or dyes.

2. The Powder

The powder or powders useful for impregnation are any that provide sufficient adsorption/and or absorption to the oil phase or phase change material and the microwave-sensitive material. Preferred are powders with large surface areas compared to the volume of the powder particles. The powder particles themselves will preferably be almost completely or substantially insensitive to microwave radiation relative to the microwave-sensitive material.

Typically, a wide range of powder particle sizes are useful with the invention. Preferred particle sizes are above about 1 micron. More preferred are particles sizes of above about 1 micron to 100 microns. Most preferred are particle sizes of about 10 to 80 microns. An important consideration is that the particles sizes selected not interfere with a high loading in the impregnation and retention of microwave-sensitive and phase-change materials on the powder. Another consideration is that larger particle sizes will reduce dust generation during the preparation of the powder.

In this regard, a most preferred powder of the type herein described is calcium silicate.

3. Preparation of the Impregnated Powder

In general it is the wax phase (in some embodiments it is solid at room temperature) of the mixture which performs much of the thermal storage function, in emulsions for compositions of thermal storage units according to the present invention. Thus, it will generally be preferred that mixtures used in thermal storage compositions according to the present invention include a relative amount, for example, by weight, of microwave-insensitive-phase-change material to microwave-sensitive material which reflects the least amount of microwave-sensitive material needed to heat the microwave insensitive-phase-change material for the particular application. The amount of phase-change material (heat storage material) desired for any given application will depend, of course, upon the size of the thermal storage unit needed; the length of time it is required to remain somewhat warm; and, the conditions under which it is to be used. Generally, impregnated powders comprising about 45 wt% to 95 wt% phase-change material, the balance comprising microwave insensitive phase, anti-microbial, and possibly other adjuvant, will be useful and preferred. The specifics given below, for an example of a seat cushion, provide basic principles of operation which can be extended to many other uses.

In one typical embodiment, it is foreseen that impregnated powders 43 according to the present invention will be prepared by mixing an emulsified melt of a microwave-sensitive material and the microwave-insensitive material together with the powder (e.g., calcium silicate powder), until all of the materials have been adsorbed and/or absorbed. Impregnation of the powder with an emulsion is preferred because the emulsion already has the microwave-sensitive materials and the phase-change materials interspersed so that the probability of obtaining the desired impregnated powder where both materials are on the same powder particle is increased.

In the method of another typical embodiment, when no emulsified mixture is used, it is contemplated that the powder can be partially adsorbed first with either the

microwave-sensitive or the microwave-insensitive material. This would be achieved by very high speed turbulent mixing as with a Pappenmeier turbulent mixer, a Henschel turbulent mixer or a Welex mixer. The remaining adsorbing capacity may then be filled in the same manner with either the microwave-insensitive or microwave-sensitive material, respectively.

In the method of yet another typical embodiment, when no emulsified mixture is used, it is contemplated that a powder can be vigorously mixed while spraying microwave-sensitive material and phase-change material, either separately or as a mixture, into the vigorously mixing powder in a manner effective to provide an impregnated free-flowing powder wherein more than at least 50 n% of all powder particles have both said microwave-sensitive material and said phase-change material impregnated therewith. It is contemplated that addition of the materials to the mixing powder be done at very high shear rates and controlled spraying so that the materials are sprayed into the vigorously-mixing turbulent powder at low volumes and preferably in small spray droplets.

In typical applications, the phase-change material useful for impregnation with the powder is heated and melted by thermal transfer from the microwave-sensitive material that is also adsorbed to the impregnated powder 43. The microwave-sensitive material may be a liquid, a solid or may itself undergo phase changes at the temperatures at which the invention is contemplated to be used (if the temperature change is large enough); however, the microwave-sensitive material is not intended to and will ordinarily not undergo phase changes. In a preferred embodiment of the invention at least 75 n% to substantially all of the impregnated powder particles have both phase-change material (oil phase) and microwave-sensitive material adhered to each powder particle. The preferred embodiment is obtained by mixing the powder with an emulsion of the phase-change material and microwave-sensitive material.

Higher costs for microwave-sensitive materials such as polyethylene glycol (which is both a microwave-sensitive material and a phase-change material in the conditions of use for the present invention) compared to the cost of paraffin waxes is one of the considerations that dictate that a combination of phase-change material and

microwave-sensitive material on the same particle is preferred. Thus, for this consideration, a large amount of paraffin wax relative to polyethylene glycol would be preferred. Another consideration, besides cost, is that by the use of two materials on the powder particles, one can tailor the thermal characteristics of the final impregnated powder to a greater degree.

Thus an additional embodiment includes a powder impregnated with a microwave-sensitive material, a phase-change material and a third material that is both an effective phase-change material and an effective microwave-sensitive material. For example, in a paraffin/water system up to 50 wt% of the material could be the third material comprising stearic acid (which is both a phase-change material and is microwave sensitive). Most preferably, only up to 25 wt% of the paraffin/water material would comprise the third material.

Arrangements where more than 50 n% to 75 n% of the powder particles have both phase-change material and microwave-sensitive material adsorbed on the same particle are workable but are less preferred. This is because heat transfer to a particle not having both materials deposited thereon depends on heat transfer from another particle that has adsorbed microwave-sensitive material. In order to have efficient heat transfer between such particles of powder, the powder particles would need to be closely packed.

A higher number of particles having both microwave-sensitive and phase-change materials absorbed thereon results in several advantages such as: (1) higher rates of heat transfer from the microwave-sensitive material to the phase-change material; (2) a looser packing of powders is possible because heat transfer to the phase-change material occurs within the same powder particle as well as between powder particles; and (3) hot or cold spots are reduced because the probability of having any given volume filled with an excess or shortage of powder particles with adsorbed microwave-sensitive material is greatly reduced. Another benefit of having both materials on each particle is that the particles can be incorporated into other materials (e.g., plastics such as shoe soles or inserts).

Each particle can then independently provide the required heating and warmth retention for the composite article.

4. Other Adjuvants

It is foreseen that in some instances other adjuvants may be provided in the mixture. For example, preservatives may be included to inhibit bacterial growth over the life of the thermal storage unit. For typical systems, for example, involving a paraffin wax/water emulsion, preservatives such as chlorobutanol; dichlorobenzyl alcohol; propylene glycol; formaldehyde; phenylmercuric acetate; benzoic acid; chloromethyl isothiazolinone; methyl isothiazolinone; dehydroacetic acid and its sodium salt; potassium sorbate; parabens; sodium pyrothione, zinc pyrothione and glutaraldehyde will be effective to accomplish this. In general, the amount of preservative should be an amount effective to achieve the desired level of resistance to biological activity. The specific formulation given hereinbelow, for a preferred arrangement usable as a seat cushion, provides further guidance with respect to this other adjuvants which may be used in thermal storage compositions according to the present invention include: dyes; antioxidants; flame retardants, etc.

5. Use of the Impregnated Powder as a Thermal Storage Composition

In use, impregnated powders according to the present invention operate as thermal storage compositions, upon activation with microwave energy. In a typical use, the impregnated powder 43 is exposed to sufficient amounts of microwave energy such that the powder and its impregnated material is heated to a temperature above the melting point of the phase-change material. In addition, sufficient energy should be imparted to the microwave-sensitive material such that by thermal transfer from the microwave-sensitive material to phase-change material, the phase-change material is melted.

Upon removal from the microwave oven, the impregnated powder 43 will be hot. It will undergo two separate steps in cooling: (1) The mass of the powder particles themselves and the mass of microwave-sensitive material adsorbed with the powder particle will ordinarily undergo sensible heat loss only; (2) The mass of phase-change material adsorbed with the powder particles will undergo three periods of cooling: a period of sensible heat loss (from the original liquid phases) above the freezing point of

the phase-change material; a period of latent heat loss during transition of the phase-change material from a liquid to solid (at about the phase transfer temperature thereof); and, a second sensible heat loss period below the melting point of the phase-change material.

The heat loss in the two periods of sensible heat loss will generally be controlled by the heat capacity of the original liquid phase-change material, microwave-sensitive material, and powder. For a liquid phase such as wax or water, the sensible heat loss will be relatively rapid by comparison to heat loss during the middle stage of transformation in the material from liquid to solid. During the stage of heat loss which occurs during the transformation of the phase-change material from a liquid to a solid, heat loss will be controlled by such characteristics as rate of crystallization of the phase-change material. Thus, it can be seen that the phase-change material serves as a thermal reservoir which will act to retain the impregnated powder composition at a relatively constant, temperature, for a significant length of time, by comparison to a system which just uses a liquid phase.

EXAMPLES

The following examples are disclosed to further teach the practice of the invention and are not intended to limit the invention as it is delineated in the claims.

Example 1

This example illustrates an embodiment where a powder is impregnated with an emulsion of a microwave-sensitive material and a phase-change material. The emulsion is a Michelman No. 30560, obtained from Michelman Inc. (Cincinnati, OH, USA), being a 65:35 wax in water emulsion (65 wt% wax) having a wax particle size of about 90 n% less than 1.6 μm, 50 n% less than 1.05 μm. The emulsion was combined in a Brabender mixer with Micro-Cel E powder at 50 rpm in a weight ratio of 3.5 parts powder to 6.5 parts emulsion at a temperature of 70°C. The mixing was at conditions that are expected to provide at least 50 n% to substantially all of the particles of the powder being

impregnated with both the microwave-sensitive material (water) and phase-change material (wax).

The Micro-Cell E is a product of Celite Corp. (Lompoc, CA, USA) and is calcium silicate powder that has a particle size distribution where 94 n% (population) of the particles are smaller than 44 μm (325 Mesh), a surface area of about 120 m^2/g, and may contain up to 2 wt% crystalline silica, i.e., quartz.

The impregnated powder product obtained was powdery and free flowing. After heating the product material to a temperature above the melting point of the wax, the material remained powdery.

Example 2

Example 1 was repeated using a weight ratio of 3.5:17 parts powder (17 wt%) to emulsion (83 wt%). A lumpy dry material was obtained. The lumpy dry material became a heavy cream (mousse like) after heating above the melting point of the wax.

Example 3

This example illustrates microwave heating of impregnated powder material with a typical 1000 watt microwave oven. Thus, 50 g of the powdered material of Example 1 was placed in a polyethylene bag and the bag placed into the oven. The oven was set at full power and the impregnated powder material reached a temperature of 54.4°C (130°F) after two minutes of heating. The powder remained a free-flowing powder after heating.

6. Specific Example of a Working Embodiment

In the following description, a working example of a specific embodiment of the present invention, involving the use of compositions for thermal storage units in the context of a seat cushion, is presented. From the specifics provided, general applications of the present invention in a wide variety of systems can be foreseen and understood.

The following materials are described for use in the context of a seat cushion for use by adults. The seat cushion described will have outer dimensions of about 32 cm x 47 cm x 3 cm.

The sheath 25 for the working example described comprises a nylon fabric (preferably a rip-stop nylon). A specific, useable material is 200 denier Nylon Oxford Taffeta, No. 68295 available from Tapetex Corp. (Rochester, NY 14623, USA). Preferably, a material is selected which has been subjected to fire-retardant treatment. This material is used for the portion of the cushion 3 corresponding to sheath 25, Fig. 3.

The foam envelope, corresponding to foam pad 26, Fig. 3, comprises any conventional seat cushion foam, for example a polyurethane foam slit to form the envelope. Preferably, fire-retardant foam is selected. A usable material is L32SX Foam, available from E.R. Carpenter (Templeton, TX 76503, USA). The foam envelope is selected with exterior dimensions substantially corresponding to those desired for the overall construction.

The thermal storage unit 20, as indicated generally above, comprises an outer pouch 40 enclosing an optional inner pouch 45, or only an outer pouch 40 which encloses impregnated powder according to the description herein. The outer pouch 40 preferably comprises an 8 mil matte two-side vinyl available under the trade name Delta 6 from Flex-Seal Packaging (Rochester, NY, USA). The dimensions of the pouch are about 30 cm x 42 cm, with a 0.6 cm heat seal along the periphery thereof. The inner pouch 45 in one embodiment preferably comprises a polymeric laminate, for example, a nylon/high density polyethylene (HDPE) polymer or a polypropylene/nylon (coated with polyvinylidene chloride) copolymer. The dimensions of the inner pouch 45 are about 28 cm x 39 cm, with a 1.25 cm heat seal around the periphery thereof. For the embodiment where the pouches are permeable to gas, a nylon fabric, preferably a rip-stop nylon as specified above for the sheath, or more preferably a polyolefin based fabric, may be used.

Received inside the inner pouch 45 (or the outer pouch 40, if only one pouch is used) is the impregnated powder 43, typically calcium silicate powder or the like, having adsorbed therewith: (1) a phase-change material that is microwave insensitive, and (2) a microwave-sensitive material. It is required that substantially all or at least 75 n%, and least preferably at least more than 50 n% (population) of the individual powder particles have both microwave-sensitive material and phase-change material present on each particle. The inner and outer pouches 40 and 45 preferably comprise a material which

is stable to the temperatures to which the system will be heated in use, does not hydrolyze under the conditions of use, and is not affected by the microwave-sensitive or insensitive material adsorbed on the calcium silicate powder.

When the powder and adsorbed emulsion or mixture is enclosed within the film of inner pouch 45 (Fig. 2) it is not necessary to apply vacuum draw to the interior of the inner pouch 45 in order to reduce air presence and facilitate close packing of the impregnated powder 43, because substantially all or at least 75 n%, and in a less preferred embodiment, more than at least 50 n%, of the particles are individually heated. The presence of microwave-insensitive-phase-change material and microwave-sensitive material on each particle provides for fast even heating of the impregnated powder and allows the impregnated powder to be loosely packed and be free to flow within the pouch much as a liquid would.

The construction described is prepared for use by placement in a 700 or 750 watt microwave oven for a few minutes on each side. It is anticipated that about 1 to 2 kg of impregnated powder would be preferred. If simply left standing at about 5°C-20°C, it would remain warm for about 4 to 8 hours. The actual rate of heat loss in use will depend, of course, upon how much of the time the cushion is used with a person sitting thereon, the size of the person, the ambient temperature and related factors.

7. Alternative Applications of Principles According to the Present Invention

The specific arrangement described above, of a seat cushion, involves an arrangement which is intended to radiate heat from an outer surface thereof to the exterior environment. Thus, the material chosen to enclose the thermal storage unit 20 is a material which will allow the thermal energy to pass toward an outer surface thereof to warm the user.

It is foreseen that in alternative embodiments, thermal storage units according to the present invention may be enclosed within insulating blankets or the like designed to retain heat therein. For example, such heating constructions might be used as hand-warming muffs, or foot-warming boots or shoes. They may also be used, for example, as thermal blankets or the like. Alternatively, as will be appreciated by those

skilled in the art, the thermal storage units can also be used for keeping materials cold. By appropriate choice of melting point of the phase-change material, colder temperatures can be maintained. Then, by cooling the composition below the melt temperature, a lower temperature will be maintained. The latter use would provide beneficial relief during hot weather, assist in food or medical storage, or provide treatment where cold compresses and the like are desired. The compositions can be kept in a regular freezer and rapidly brought to the desired application temperature for use either as a heating or cooling medium.

Another alternative embodiment includes a thermal storage composition where a calcium silicate powder is only impregnated with a material that is both a microwave-sensitive material and a phase-change material. This embodiment would likewise be a free-flowing powder.

While the forms of the invention herein disclosed constitute presently preferred embodiments, many others are possible. It is not intended herein to mention all of the possible equivalent forms or ramifications of the invention. It is to be understood that the terms used herein are merely descriptive, rather than limiting, and that various changes may be made without departing from the spirit of the scope of the invention.

WHAT IS CLAIMED IS:

1. A thermal storage composition activatable by exposure to microwave energy, said composition comprising a powder wherein more than at least 50 n% of all particles of said powder are impregnated with both a phase-change material and a microwave-sensitive material.

2. A thermal storage composition according to Claim 1, further comprising a third material that is both an effective phase-change material and microwave-sensitive material.

3. A thermal storage composition according to Claim 1, wherein at least 75 n% of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

4. A thermal storage composition according to Claim 1, wherein substantially all of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

5. A thermal storage composition according to Claim 1, wherein said thermal storage composition is a free-flowing impregnated powder.

6. A thermal storage composition according to Claim 1, wherein said phase-change material comprises microwave-insensitive material.

7. A thermal storage composition according to Claim 1, wherein said microwave-sensitive material comprises a low volatility, microwave-sensitive glycol.

8. A thermal storage composition according to Claim 7, wherein said phase-change material comprises organic wax material.

9. A thermal storage composition according to Claim 1, wherein said phase-change material comprises wax selected from the group consisting essentially of: paraffin wax; beeswax; candelilla wax; carnauba wax; cotton wax; wool wax; montan wax; and, mixtures thereof.

10. A thermal storage composition according to Claim 1, wherein said phase-change material comprises material having a melting point of at least 30°C.

11. A thermal storage composition according to Claim 1, wherein said phase-change material comprises material having a melting point within the range of 35°C-65°C.

12. A thermal storage composition according to Claim 11, wherein said powder comprises calcium silicate, said phase-change material comprises paraffin wax and said microwave-sensitive material is selected from the group consisting of a microwave-sensitive glycol, water and mixtures thereof.

13. A thermal storage composition according to Claim 11, wherein said powder comprises calcium silicate, said phase-change material comprises paraffin wax and said microwave-sensitive material is selected from the group consisting of dipropylene glycol, diethylene glycol, higher oligomers of propylene or ethylene glycol, water, and mixtures thereof.

14. A thermal storage composition activatable by exposure to microwave energy, said composition comprising:

(a) a powder; and

(b) an emulsion comprising:

(1) a microwave-sensitive material, and

(2) a phase-change material; and,

wherein said powder is impregnated with said emulsion.

15. A thermal storage composition according to Claim 14, wherein at least more than 50 n% of all particles of said powder are impregnated with both a phase-change material and a microwave-sensitive material.

16. A thermal storage composition according to Claim 14, wherein at least 75 n% of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

17. A thermal storage composition according to Claim 14, wherein substantially all of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

18. A thermal storage composition according to Claim 14, wherein said impregnated powder is free-flowing at temperatures of use.

19. A thermal storage composition according to Claim 14, wherein said powder comprises calcium silicate.

20. A thermal storage composition activatable by exposure to microwave energy, said composition a product of a process comprising:

(a) emulsifying a phase-change material and a microwave-sensitive material; and

(b) impregnating a powder with said emulsion in an amount effective to form a free-flowing impregnated powder.

21. A process for producing a thermal storage composition comprising:

(a) emulsifying a mixture of a phase-change material and a microwave-sensitive material; and

(b) mixing said emulsion with a powder able to be impregnated with said emulsion in an amount wherein said thermal storage composition is a free-flowing impregnated powder.

22. A process for producing a thermal storage composition comprising:

(a) mixing a microwave-sensitive material or a phase-change material with a powder able to be impregnated with said materials, in an amount, at a temperature, and at a mixing shear rate effective to impregnate said microwave-sensitive material or said phase-change material with said powder particles to form a partially impregnated powder able to receive additional microwave-sensitive material or phase-change material; and

(b) mixing microwave-sensitive or said phase-change material not selected in step (a) with said partially impregnated powder in an amount, at a temperature, and at a mixing shear rate effective to impregnate said microwave-sensitive or said phase-change material with said partially impregnated particles, wherein a product is obtained with at least more than 50 n% of said particles having both said microwave-sensitive material and said phase-change material impregnated therewith.

23. A process according to Claim 22, wherein at least 75 n% of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

24. A process according to Claim 22, wherein substantially all of said powder particles have both said microwave-sensitive material and said phase-change material impregnated therewith.

25. A process for producing a thermal storage composition comprising:

(a) vigorously mixing a powder;

(b) spraying microwave-sensitive material and phase-change material, either separately or as a mixture, into the vigorously mixing powder in a manner effective to form an impregnated free-flowing powder wherein at least more than 50 n% of all powder particles have both said microwave-sensitive material and said phase-change material impregnated therewith.

26. A process according to Claim 25, wherein at least 75 n% of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

27. A process according to Claim 25, wherein substantially all of said powder particles have both said microwave-sensitive material and said phase-change material impregnated therewith.

28. A thermal storage unit which is activated by exposure to microwave energy; said thermal storage unit comprising:

(a) a microwave-transparent container; and,

(b) a thermal storage composition enclosed within said container; said thermal storage composition comprising:
a powder impregnated with a microwave-sensitive material and a phase-change material, wherein more than 50 n% of all particles of said powder are impregnated with both a phase-change material and a microwave-sensitive material.

29. A thermal storage unit according to Claim 28, wherein said microwave-sensitive material comprises water; and, said phase-change material comprises paraffin wax.

30. A thermal storage unit according to Claim 28, wherein said container comprises a flexible pouch.

31. A thermal storage unit according to Claim 28, wherein said impregnated powder was impregnated with an emulsion.

32. A thermal storage unit according to Claim 28, wherein substantially all particles of said powder are impregnated with both a phase-change material and a microwave-sensitive material.

33. A heating construction comprising:

(a) a microwave-transparent outer cover; and,

(b) a thermal storage unit activatable by exposure to microwave energy; said thermal storage unit being positioned within said outer cover; said thermal storage unit comprising:

(1) a microwave-transparent outer pouch; and,

(2) an impregnated powder composition within said outer pouch; said impregnated powder composition comprising: a powder impregnated with a microwave-sensitive material and a phase-change material, wherein at least more than 50 n% of all particles of said powder are impregnated with both a phase-change material and a microwave-sensitive material.

34. A heating construction according to Claim 33, wherein said microwave-sensitive material comprises water; and said phase-change material comprises paraffin wax.

35. A heating construction according to Claim 33, wherein said impregnated powder was impregnated with an emulsion.

36. A heating construction according to Claim 33, wherein said microwave-transparent outer cover comprises a seat cushion.

37. A process of storing thermal energy for release over a extended period of time; said process including a step of:

(a) exposing a powder composition to microwave energy; said powder composition comprising:

(1) a powder; and,

(2) a microwave-sensitive material and a phase-change material impregnated with said powder, wherein at least more than 50 n% of all particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material;

(b) said step of exposing said powder composition to microwave energy comprising exposing said powder composition to microwave energy of appropriate power and for a sufficient period of time to heat said powder composition to a temperature above the melting point of said phase-change material and to melt said phase-change material.

38. The process of Claim 37, wherein substantially all of said particles of said powder are impregnated with both said phase-change material and said microwave-sensitive material.

39. A thermal storage composition activatable by exposure to microwave energy, said composition comprising:

a powder comprising calcium silicate impregnated with a material that is both an effective microwave-sensitive material and a phase-change material.

40. All novel disclosures and combinations thereof.

FIG. 1

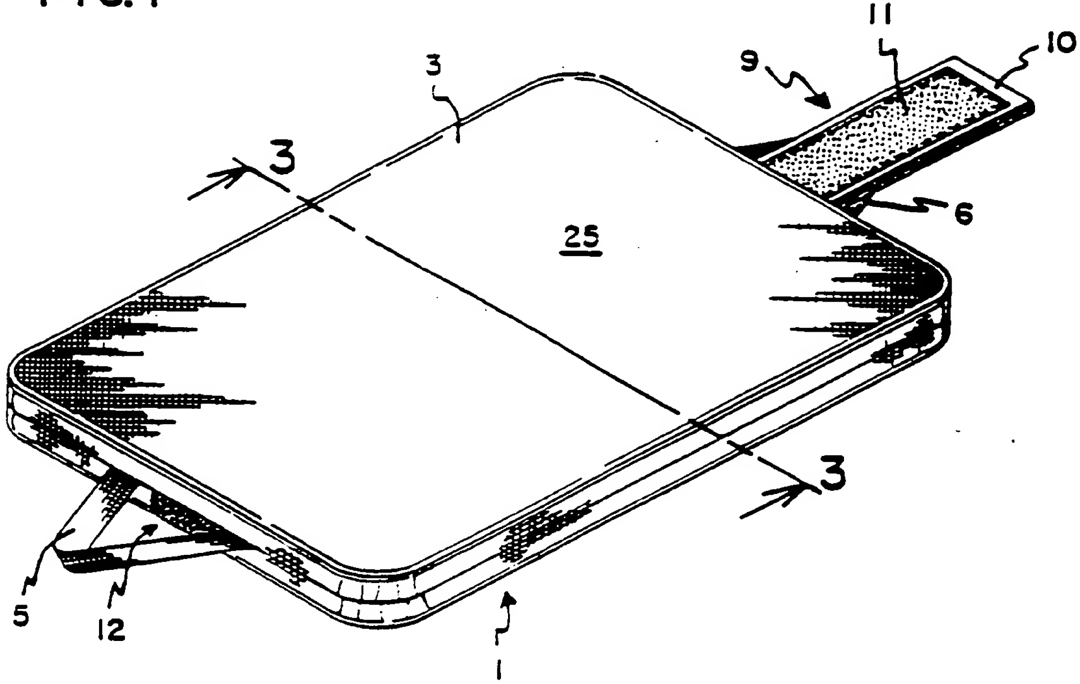
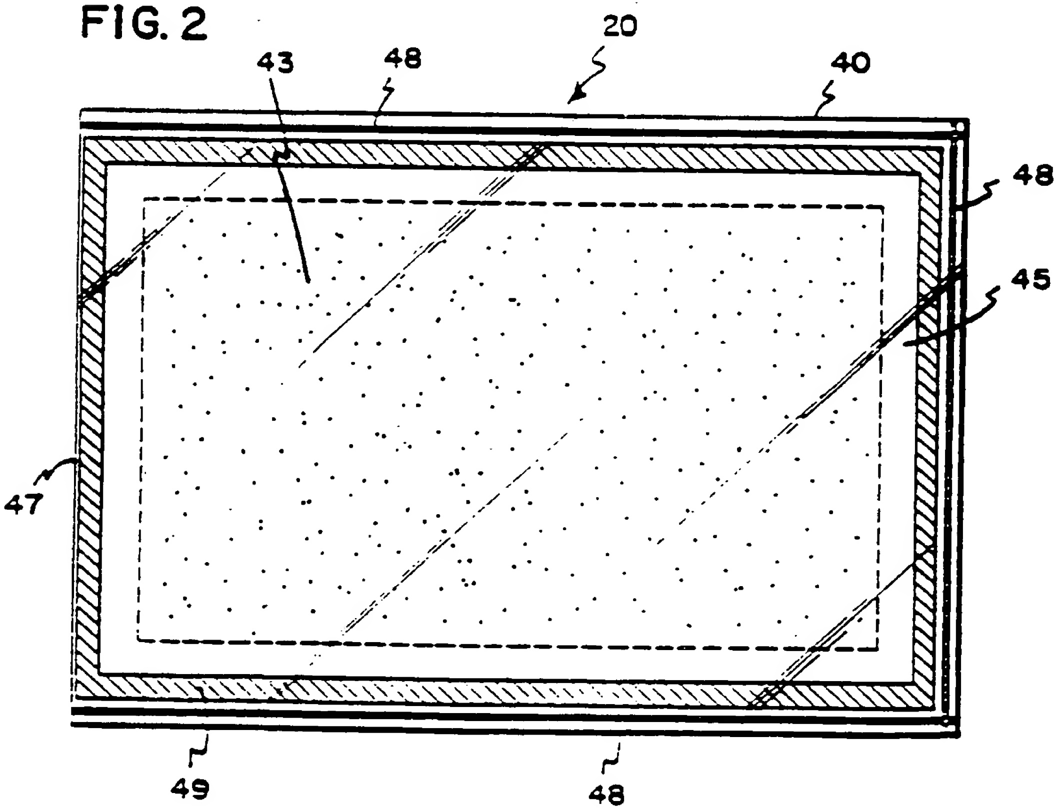


FIG. 2



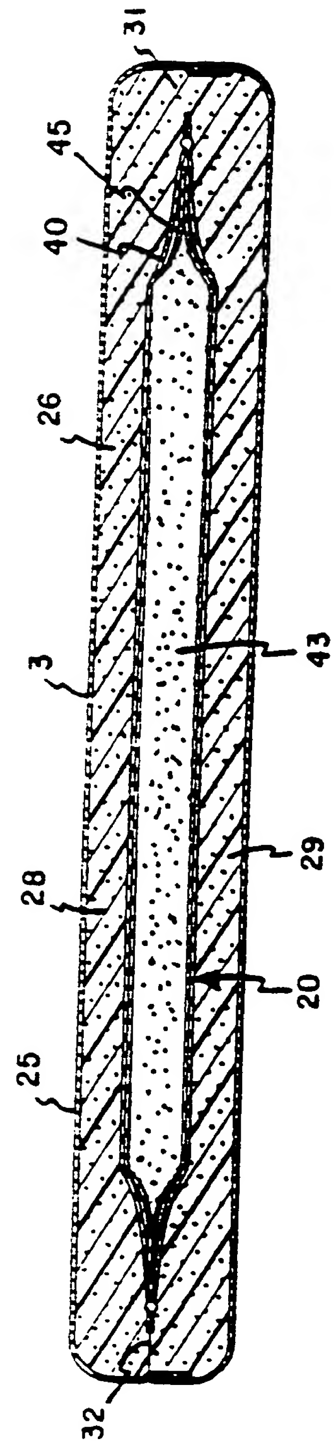


FIG.3

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US96/06685

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :H05B 6/80; A61F 7/00.
US CL :219/759; 607/101, 108, 114; 427/213.3; 126/204.
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 219/759, 730; 607/96, 101, 102, 108-112, 114; 126/204; 252/71; 427/213.3, 214, 595.

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	JP, A, 1-123917 (NOK CORP) 16 MAY 1989 See the English Abstract.	1-6 and 9 --------- 7, 8 and 10-39
X - - Y	JP, A, 4-39381 (NIPPON OIL KK) 10 FEBRUARY 1992 See the English Abstract.	1-6 and 9 ---------- 7, 8 and 10-39
Y	JP, A,2-302489 (YAMAGATA) 14 DECEMBER 1990 See the English Abstract.	1-39
Y	US, A, 5,300,105 (OWENS) 05 APRIL 1994 See Figures 1, 8 and 9.	33-36
Y	US, A, 4,849,593 (HUGHES ET AL) 18 JULY 1989 See Figures 11 and 12.	33-36

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
23 JULY 1996	17 SEP 1996

Name and mailing address of the ISA/US	Authorized officer
Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	PHILIP H. LEUNG
Facsimile No. (703) 305-3230	Telephone No. (703) 308-1782

Form PCT/ISA/210 (second sheet)(July 1992)★

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 5,070,223 (COLASANTE) 03 DECEMBER 1991 See Figures 1-5 and col. 5, line 10 - col. 6, line 53.	33-36
Y	US, A, 5,202,150 (BENSON ET AL) 13 APRIL 1993 See the entire document.	21-27, 37 and 38
A, P	US, A, 5,424,519 (SALEE) 13 JUNE 1995 See the Entire document.	1-39

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US96/06685

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. [X] Claims Nos.: 40
because they relate to subject matter not required to be searched by this Authority, namely:

There is no definite claimed structure in the claim and the scope therefore is not known.

2. [ ] Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. [ ] Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. [ ] As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. [ ] As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. [ ] As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. [ ] No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest
[ ] The additional search fees were accompanied by the applicant's protest.
[ ] No protest accompanied the payment of additional search fees.